Laboratory Name: Sandia National Laboratories, CA
B&R Code: KC020101

FWP and possible subtask under FWP:
FWP Alloy Theory

FWP Number: SCW4341

Program Scope:
The Alloy Theory program at Sandia, California is focussed on the development, validation and application of computational methods that can be used to explain and predict structural, mechanical, and thermodynamic properties of materials. A technical thrust is on the development and application of parameter-free first-principles methods. Quantum mechanical calculations are used to obtain structural energies, and statistical physics simulations are used to account for thermal effects. These techniques are tested by direct comparison to experimental measurements both in our group as well as elsewhere. Therefore, this program is highly linked to an experimental effort that is supported under SCW 604. The ultimate goal of the program is to uncover fundamental concepts that can be applied to understanding the structure, behavior and processing of alloys. The main research areas are the following: (i) non-configurational (vibrational, electronic, and magnetic) free energies in bulk alloys, (ii) structure of and ordering in ultrathin alloy films, and (iii) structure and thermodynamics of liquid alloys.

Major Program Achievements (over duration of support):
Key accomplishments include: demonstrating a 27-fold increase in the solid solubility of Sc in Al due to vibrational entropy effects, theoretical discovery of vibrational stabilization of the phase of Al_2Cu in the classical Al-Cu system, explanation of magnetic Invar properties of Fe-Ni quantitative experimental and theoretical study of short-range order in Pd-Au surface alloys on Ru(0001), prediction and experimental verification (supported under SCW 604) of an unusual atomic structure in Co-Ag surface alloys on Ru(0001), *ab-initio* molecular dynamics study of liquid Ni-Al, and development of accurate methods for calculating anisotropy of solid-liquid interfacial free energy.

Program Impact: Has expanded our understanding of the mechanisms solute stability in alloys and their implications to alloy behavior.

Interactions:
Prof. M. Asta (Dept of Materials Science, Northwestern University, Evanston, Illinois)
Dr. J. J. Hoyt (Sandia National Laboratories, Albuquerque, New Mexico)
Prof. D. Seidman (Dept of Materials Science, Northwestern University, Evanston, Illinois)
Dr. C. Wolverton (Ford Motor Laboratories, Dearborn, Michigan):

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
M. Asta organized the Workshop on Thermodynamic and Structural Properties of Alloy Materials held on 20-24 June, 1999 in Oranjestad, Aruba. This workshop was dedicated to Professor Didier de Fontaine of the University of California at Berkeley.

V. Ozolins is one of the organizers of the next Workshop on Thermodynamic and Structural Properties of Materials, to be held in 2003.

Personnel Commitments for FY2002 to Nearest +/- 10%:
V. Ozolins (100%), R.Q. Hwang (50%),

Authorized Budget (BA) for FY00, FY01, FY2002:
FY00 BA $352k **FY01 BA $**342k **FY02 BA $**349k

Laboratory Name: Sandia National Laboratories, CA
B&R Code: KC020101

FWP and possible subtask under FWP:
FWP Surface, Interface and Bulk Properties of Advanced Ceramics

FWP Number: SCW1550

Program Scope:
This project focuses upon the science of ceramic surfaces and their interfaces with metals. Our effort includes determining the surface structure of ceramics such as alumina using electron diffraction analysis. In addition, we directly follow the dynamics of surface morphology and structure in real time using the microscopic techniques of scanning tunneling microscopy (STM) and low-energy electron microscopy (LEEM). We strive to quantitatively understand how mass transport occurs on the surface and through the bulk and to characterize the nature and energetics of the bulk thermal defects. We form metal/ceramic interfaces by depositing metals on ceramic single crystals (such as alumina and titanium dioxide) and by oxidizing metal alloys (such as NiAl). We determine how the metals nucleate and grow on ceramic surfaces and the mechanisms of alloy oxidation. We seek to understand how metal/ceramic interfaces evolve during film growth and how interface structure, bonding, and reaction influence the interfacial energy ("strength") and microstructure of the growing film. Through understanding the energetics of metal particles on ceramics, we hope to be able to control the size and morphology of the particles. Our long-range goal is to develop sufficient understanding so ceramic surfaces and their interfaces with metals can be scientifically tailored for improved performance and new properties.

Major Program Achievements (over duration of support):
Key accomplishments include: Discovered the "self-limiting" growth of Cu and Ag particles on the TiO_2 (110) surface; Determined the structure of the α-Al_2O_3 (0001) surface; Developed a new technique to determine the nature and energetics of bulk defects; Showed that bulk diffusion (rather than surface diffusion) can dominate the morphological evolution of a surface; Observed how a binary alloy oxidizes to form alumina.

Program Impact: Have applied novel electron and scanning probe microscopies to the investigation of oxides leading to the discovery of new mechanisms coupling the bulk to the surface and interfaces of oxides.

Interactions:
Dr. M.A. Van Hove, LBNL
Dr. D.L. Medlin, Sandia National Labs
Dr. N.C. Bartelt, Sandia National Labs
Dr. D. Jennison, Sandia National Labs

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
K.F. McCarty: Invited talk at 2001 American Vacuum Society Meeting, invited talk at the 2002 ACS meeting

Personnel Commitments for FY2002 to Nearest +/- 10%:
K.F. McCarty (100%)

Authorized Budget (BA) for FY00, FY01, FY2002:
FY00 BA $392k **FY01 BA $**399k **FY02 BA $**359k

Laboratory Name: Sandia National Laboratories, CA
B&R Code: KC020102

FWP and possible subtask under FWP:
FWP Science of Materials, subtask on Transitions in the Strongly Collective Behavior of Dislocations and Interfaces at the Onset of the Nanoscale Regime

FWP Number: SCW604

Program Scope:
We use microstructure evolution during deformation as a probe of fundamental properties. Quantitative analysis and measurements in the TEM of structures that are created during monotonic deformation from small to large strains have shown that the structures maintain a similar character with increasing strain. Measurements include the averages and distributions of boundary misorientation angle/axis pair, inclination plane, spacing, and width for two types of deformation induced dislocation boundaries. Scaling of the distributions of these features and similitude provide governing principles for structure evolution. The development of these structures has been quantitatively linked to strength increases using an additive law for dislocation strengthening by low angle boundaries and a Hall-Petch type strengthening due to high angle boundaries. Theoretical modeling of this evolution is therefore underway including the modeling of the spacing distributions and a multiscale coupling of three-dimensional dislocation dynamics and continuum plasticity. Emphasis is placed on the boundary spacing both experimentally and theoretically since modeling the spacing evolution is the missing link. The lower spatial limits of this behavior are being explored using graded nanostructures produced by sliding deformation. This new approach has shown a remarkable invariance on the pattern of boundary spacings with average sizes from 10,000 to 10 nm. This universal scaling behavior indicates a continuity in the processes that create and remove boundaries with increasing deformation. Exploration of the regime below 10nm is the next step.

Major Program Achievements (over duration of support):
Key accomplishments include: Discovered that probability distributions of dislocation boundary spacings remains the same when normalized by the average spacing for a wide strain range, different materials and deformation conditions; Established the same scaling behavior of boundary spacings is observed from 10,000 to 10nm indicating the same fundamental deformation processes of dislocation slip control the evolution over this range; Geometric and analytical models for the evolution of spacing probability distributions; Developed stereological methods to measure spacing and probability distributions for the spacings of different types of deformation-induced dislocation boundaries; Created technique to explore deformation behavior from 10,000 to 10nm with graded microstructures produced by sliding; Established the role of microstructure in crystallographic texture formation; Simulated planar dislocation boundaries using a multi-scale coupling of a 3-D discrete dislocation dynamics code with continuum finite element code; Predicted for the first time flow stress, from yield to large strains, using the measured microstructure parameters and a new model based on i) dislocation strengthening due to the presence of low angle boundaries and ii) grain boundary strengthening due to medium and high angle boundaries.

Program Impact: Have discovered and investigated new scaling concepts that govern deformation structure over a wide range of strains, materials and deformation conditions.

Interactions:
Prof. H. Zbib, Wash. State Univ.
Dr. Niels Hansen, Risø National Laboratory in Denmark

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
D.A. Hughes: Editorial Board of International Journal of Plasticity, Organized symposium for MRS spring 2001, Orgnanized symposium for Plasticity 1999, Best poster MRS Spring 2001
10 Invited talks since 2000

Personnel Commitments for FY2002 to Nearest +/- 10%:
D.A. Hughes (100%), S. Aubry (postdoc) 100%

Authorized Budget (BA) for FY00, FY01, FY2002:
FY00 BA $430k **FY01 BA $**360k **FY02 BA $**357k

Laboratory Name: Sandia National Laboratories, CA
B&R Code: KC020102

FWP and possible subtask under FWP:
FWP Science of Materials, subtask on Grain Boundary Structure and Dynamics

FWP Number: SCW604

Program Scope:
We are investigating the structure and dynamical behavior of grain boundaries through a coupled program of experimental and theoretical methods. Our experimental effort utilizes both conventional and high-resolution transmission electron microscopy, to probe the elements of interfacial structure, and *in situ* measurements, to directly investigate the dynamic processes of interfacial motion and dislocation-grain boundary interaction. In our experiments, we investigate systems of carefully controlled crystallographic geometry, fabricated either through bulk bicrystal methods or through thin film epitaxial growth. Our theoretical work combines continuum elasticity, interfacial crystallography, atomistic simulations, and first-principles computations. We are using both semiempirical methods, such as the embedded atom method, and newer state-of-the-art first principles and transition state finding techniques. The semiempirical approaches allow rapid qualitative testing of concepts in grain boundary dynamics. For first principles calculations, we are using VASP, which allows quantitative calculations of grain boundary properties. For example, we have used VASP and transition state finding to calculate the ideal shear strength of an aluminum =3 {112}boundary. These calculations also reveal the coordinated atomic displacements associated with grain boundary migration coupled to grain boundary sliding. Throughout, our approach is to employ both experimental and theoretical tools to obtain a basic scientific understanding of the fundamental structural elements, interactions and excitations that govern grain boundary behavior.

Major Program Achievements (over duration of support):
Key accomplishments include: Developing an extended Peierls-Nabarro model for dislocations based on *ab initio* Local-Density Approximation; Observation and modeling of the core structure of dissociated $\frac{1}{3} < \mathbf{111} >$ twin dislocations; Simulation of aluminum grain boundary sliding; Investigations of the influence of dislocation structure on the 9R reconstruction at Σ=3 grain boundaries; Developing a dislocation-based model for grain boundary dissociation; Multi-scale modeling of grain boundary de-faceting phase transition; Investigations of the origin of dislocations at grain boundary facet junctions

Program Impact: Have developed a quantitative understanding based on coupling simulation and microscopy of grain boundary dislocation structures and their mechanisms of motion in response external forces.

Interactions:
Prof. C.B. Carter, University of Minnesota
Prof. C.L. Briant, Brown University
Dr. G.H. Campbell, LLNL
Dr. E. Stach, LBNL-NCEM
Dr. S.M. Foiles, SNL/NM

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
D.L. Medlin Organizer of symposium at the Fall 2000 Meeting of the Materials Research Society, Council Member (since 2000) Northern California Society for Microscopy, Organizer symposium on "Thin Film Microsturcure Analysis" to be held at the Scanning 2002 meeting, May, 2002.
5 Invited talks since 1999

Personnel Commitments for FY2002 to Nearest +/- 10%:
D.L. Medlin (100%), J.C. Hamilton (50%), G.A. Lucadamo (postdoc) 100%

Authorized Budget (BA) for FY00, FY01, FY2002:
FY00 BA $385k **FY01 BA $**369k **FY02 BA $**385k

Laboratory Name: Sandia National Laboratories, CA
B&R Code: KC020102

FWP and possible subtask under FWP:
FWP Science of Materials, subtask on Surface Dynamics

FWP Number: SCW604

Program Scope:
The goal of this project is to quantify the fundamental processes governing the dynamics of surface structure and morphology. We use state-of-the-art microscopy (LEEM and STM) to measure, often in real time, the time evolution of surface structure. We use these measurements to write down precise equations of motion to describe the observed time dependence. We relate these equations of motion to atomic processes. We have used this general approach on a variety of different problems in surface science. We have studied, for example, the kinetics of surface alloying (Sn/Cu(111)), misfit dislocation dynamics (Cu/Ru(0001), 2-D grain coarsening (O/Ru(0001), 2-D self-assembly (S/Ag/Ru(0001), Pb/Cu(111)), and thermal surface smoothing (NiAl(110)). This work has revealed unanticipated mechanisms of surface motion. The goal of future work will be to determine how general these mechanisms are, as well as to further develop the conceptual framework needed to account for our observations. The ultimate goal of this work is to provide the groundwork for the quantitative, predictive capabilities needed to engineer surface properties.

Major Program Achievements (over duration of support):
Key accomplishments include: Studying the dynamics of surface alloy formation to elucidate the stability and formation of novel surface alloys and their structure. Investigating the dynamics governing self-assembly of nanoscale patterns on surfaces. Quantitative measurements of dislocation dynamics in thin film and their relationships to surface morphology, chemical reactivity and alloying. Dynamics of surface morphology and the quantitative link between bulk vacancy formation and transport to surface smoothing.

Program Impact: Have developed a quantitative understanding of many of the key dynamic processes that govern surface morphology, atomic structure and thin film growth.

Interactions:
Dr. Doon Gibbs, Brookhaven National Lab
Dr. Jan Hrbek, Brookhaven National Lab
Dr. David Zehner, Oakridge National Lab
Prof. C. Barry Carter, University of Minnesota
Prof. Shirley Chiang, University of California, Davis
Dr. James B. Hannon, IBM Research
Dr. Gary Kellogg, Dr. Peter Feibelman, Dr. Brian Swartzentruber, Dr. Jack Houston, Dr. Jerry Floro, Sandia National Labs

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
Norman C. Bartelt, 2001 MRS Medal
Gayle E. Thayer, 2001 Nottingham Prize
Robert Q. Hwang: Conference Co-chair, 2000 Spring MRS meeting

Personnel Commitments for FY2002 to Nearest +/- 10%:
N.C. Bartelt 100%, J.C. Hamilton 50%, R.Q. Hwang 50%, G.E. Thayer (student) 100%, W. Ling (postdoc) 100%, K. Thuermer (postdoc) 100%

Authorized Budget (BA) for FY00, FY01, FY2002:
FY00 BA $578k **FY01 BA $**554k **FY02 BA $**701k